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TREATMENT OF WASTEWATER FROM CAR WASH BY FENTON AND PHOTO-FENTON OXIDATIVE PROCESSES

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Abstract

Car wash generates wastewater that contains several residues, such as gasoline, lubricating oil and grease, as well as chemicals used for vehicle hygiene. The aim of this study was to investigate the application of advanced Fenton and photo-Fenton oxidative processes to treat contaminated car wash wastewater. The samples were collected from a car wash facility located in the city of Florianópolis, Santa Catarina, Brazil. Continuous flow experiments were performed, and comparisons were made between the Fenton and photo-Fenton methods. Treatment efficiency was examined through physical analysis (color and turbidity) and chemical analysis (chemical oxygen demand, surfactant, oils, and grease). Photo-Fenton treatment achieved approximately 93% of chemical oxygen demand removal, whereas Fenton treatment reached 83%. Other parameters examined showed a reduction between 75.0% and 99.6%, which means that they can be used either as an alternative method or in association with the conventional techniques for the treatment of car wash wastewater effluents.

Keywords: Effluent; Car wash; Fenton; Photo-Fenton.

1. Introduction

Filling stations are service providers that sell fuels to motor vehicles throughout the country. Car wash is an additional service offer to customers. This activity generates 150 to 350 liters of wastewater per vehicle that contain a large variety of pollutant effluents and residues of gasoline, lubricating oil and grease, as well as chemicals used for vehicle hygiene [1-3].

Nomenclatures

pH Hydrogen-ion activity

Abbreviations

ABNT	Brazilian Association of Technical Standards
ASTM	American Society for Testing and Materials
BOD	Biochemical oxygen demand
COD	Chemical oxygen demand, mg/L
CONAMA	National Environmental Council
MDF	Medium-density fibreboard
NBR	Brazilian Regulatory Standard
QWC	Queensland Water Commission
UV	Ultraviolet radiation, nm
UVA	Ultraviolet radiation A, nm
VIS	Visible

Some of these substances are considered to be recalcitrant, and when discharged into water bodies cause irreparable damage to aquatic flora and fauna, given that they present, for the most part, high potential of toxicity and bioaccumulation capacity. They can also interfere with gas exchange and energy transfer, and may affect human health. One of the main environmental problems caused by these pollutants is the decrease in oxygen concentration in the discharged effluent, which may have several environmental implications, such as fish mortality [1, 2, 4].

Because hydrocarbons are immiscible in water and due to their pollutant potential, as well as the presence of surfactants in car wash wastewater, treatment prior to final disposal should not be neglected. Most countries have issued a number of laws on wastewater recycling associated with car wash [2, 5].

To regulate the car wash activity, and in order to avoid the risks to the diverse ecosystems, the Brazilian legislation requires environmental licensing by the responsible companies. At the national level, the Resolution N.º 273 of the National Environmental Council issued in 2000 classifies gas stations and services as potentially or partially polluted enterprises [6]. So, a compulsory license is required from them. CONAMA Resolution N.º 430/2011 provides on the conditions and standards for the discharge of effluents [7]. The Brazilian Association of Technical Standards regulates the implementation of car wash systems through ABNT NBR 15594-6: 2013, part 6, which provides on the operation and maintenance of car washing [8]. In the state of Santa Catarina, the Environmental Foundation is the legal body responsible for licensing.

Global environmental legislation and guidelines on this issue are in evidence, but they are not always sufficient. In Queensland, Australia, it is mandatory to use a maximum of 70 L of fresh water in a single-car wash, and in Europe, some countries restrict water consumption to 60-70 L per car and/or impose a reclamation percentage as high as 70-80% [5, 9-12].

Currently, in Brazil, primary treatment processes are adopted that remove suspended oils and sedimentable solids. However, the system is deficient to remove parameters such as chemical demand for oxygen and surfactants. To complete the treatment, the effluents should be directed to a sanitary sewage system. If the pollutant parameters are high, its treatment can become very expensive. Therefore, to treat such effluents where they are generated is essential to protect the ecosystem [13-16].

In view of the limitation presented by the primary treatment system, an association between this method and other technologies, such as advanced oxidative processes, is required [4, 5, 17-19]. Research has been carried out on treatment methods, such as ozonation [9] and biological-based methods [20] for the treatment of car wash effluents. Ozonation leads to the formation of undesirable by-products, such as aldehydes, ketones, and carboxylic acids [21], whereas the biological treatment method is a potential area of concern due to the high COD/BOD ratio and nutrient deficiency in relation to macro and micronutrients. Hence, wastewater can delay or hold back the effectiveness of biological processes [20, 22]. However, the Fenton and photo-Fenton processes are noteworthy for their operational simplicity and high degradation efficiency [5, 23-28]. Such processes allow for the mineralization of numerous resistant substrates, that is, the organic pollutants are converted to carbon dioxide and water.

In the Fenton reaction, the hydroxyl radical generation is made by the decomposition of hydrogen peroxide (H_2O_2) catalysed by bivalent iron (Fe^{2+}) in acidic medium (H^+). The ferric ions formed can decompose H_2O_2 into peroxide radicals ($\cdot\text{OH}$), which provide the oxidation of organic and inorganic compounds present in the effluent [24]. One of the advantages of this method is that excess hydrogen peroxide in the medium decomposes into water (H_2O) and oxygen (O_2) [23, 25, 29].

Photo-Fenton treatment results from the combination of Fenton reaction with ultraviolet radiation. UV radiation can be induced by light bulbs or even by sunlight [4, 5, 30].

This study evaluated comparatively the advanced Fenton and photo-Fenton oxidative processes as an alternative to remove the parameters of color, turbidity, chemical oxygen demand, surfactants, and oils and grease from car wash effluents.

2. Experimental Procedure

The experimental procedure was divided into sampling, development of the Fenton and photo-Fenton processes and determination of the physical parameters (color and turbidity) and chemical parameters (chemical oxygen demand, surfactants, and oils and greases) from car wash effluents.

2.1. Sampling and Fenton and photo-Fenton processes

Two samples of car wash wastewater were collected before undergoing physical treatment twice within a 24-hour interval, on May 4 and 5, 2015, at a gas station located in Itacorubi, Florianópolis, state of Santa Catarina, Brazil. The samples

were analysed within a maximum 48-hour period before and after undergoing Fenton and photo-Fenton oxidative process [31, 32].

The system was assembled using a glass container for water circulation, a glass bulb with two openings, an MDF box, a magnetic stirrer, two 40W fluorescent black light bulbs (UVA radiation), a submersible pump, and hoses. All experiments were run continuously using 200 mL samples, which were adjusted to pH 3.0 with dilute aqueous sulfuric acid solution, followed by the addition of 3.0 mL of hydrogen peroxide (30% v/v) and 0.6 g of ferrous sulfate heptahydrate. The samples remained circulating for 1 hour for each process [33]. Figure 1 displays the sample circulation system used in the Fenton and photo-Fenton processes. In Fig. 1(a), the MDF box is closed. In Figs. 1(b) and 1(c), the MDF box is open to visualize the lamps. For the Fenton system, the lights have been switched off (1b, lights off), whereas for the photo-Fenton system the lights remained on (Fig. 1(c), lights on).

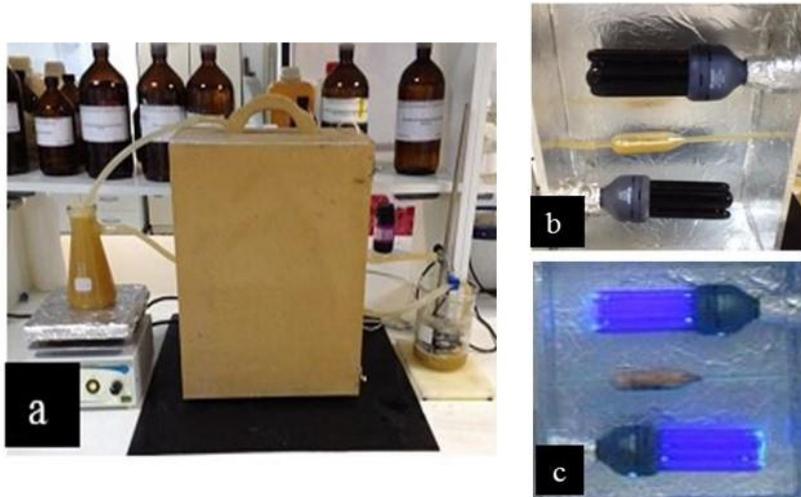


Fig. 1. (a) Fenton and photo-Fenton circulation system; (b) black lights off (Fenton), and (c) black lights on (photo-Fenton).

Source: Adapted from Ballmann, 2011 [33].

The photo-Fenton process was conducted under identical reaction conditions used in the Fenton process; however, radiation from two compact fluorescent lamps was added (Fig. 1). The sample passed between the lamps. After the sample circulation, the hydrogen ion potential was adjusted to 6.5 with the addition of 45 mL of 0.10 M sodium hydroxide. After 1-hour precipitation in an Imhoff cone, physical analysis (color and turbidity) and chemical analysis of the effluent (hydrogen ion potential, surfactants, oils and greases, and chemical oxygen demand) were carried out. The values found were compared with those collected before the Fenton and Photo-Fenton processes.

2.2. Color

ASTM D1209-05 (2011) was used as a standard test method for color measurement [34]. The Hach Model CO-1 manually-operated colorimeter was

used for color analysis. The reading was done in triplicate, and the mean values were presented.

2.3. Turbidity

For turbidity determination, we used an ALFAKIT AT 2K turbidimeter, calibrated with distilled water. The sample was added and reading was done in triplicate, with the result being the average values obtained [35].

2.4. Hydrogen-ion potential

To determine the hydrogen-ion potential, the analysis was performed at the time of collection using a Toledo SG23 Metmeter (SevenGo Duo) previously calibrated with a solution of pH 7.01 and pH 4.0 [36].

2.5. Surfactants

For the determination of surfactants, we used the ASTM D1681-05 (2014) [37] and ABNT NBR 10738 (1989), procedure A [38]. The absorbance was obtained by using a UV/VIS spectrophotometer at 652 nm. The calibration curve was plotted using a standard linear alkyl sulfonate solution in water. Figure 2 shows the schematic diagram for the determination of surfactant concentration.

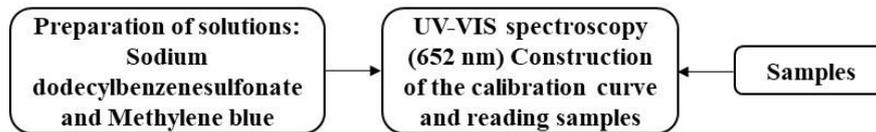


Fig. 2. Schematic diagram for the determination of surfactant concentration.

2.6. Oils and greases

Oils and greases were determined according to the ASTM D3921-96 (2011) [39] and ABNT NBR 13348 (2016) guidelines [40]. Figure 3 displays the schematic diagram for the determination of oil/grease concentration.

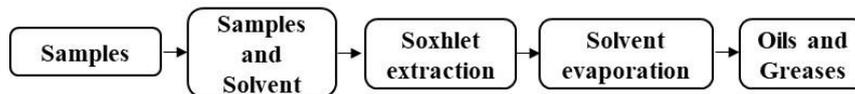


Fig. 3. Schematic diagram for the determination of oil and grease concentration.

2.7. Chemical oxygen demand

The chemical oxygen demand was determined according to the ASTM D1252-06 (2012) [41] and ABNT NBR 10357 (1988) guidelines [42]. The closed reflux method is more economical in the use of reagents and generates smaller quantities of hazardous waste, but requires homogenization of samples containing suspended solids to obtain reproducible results. Figure 4 exhibits a schematic diagram for COD determination.

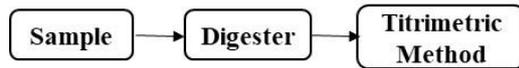


Fig. 4. Schematic diagram for COD determination, using a closed reflux method.

3. Results and Discussion

Figure 5 shows the crude sample (5a) and the samples after Fenton treatment (5b) and photo-Fenton treatment (5c).

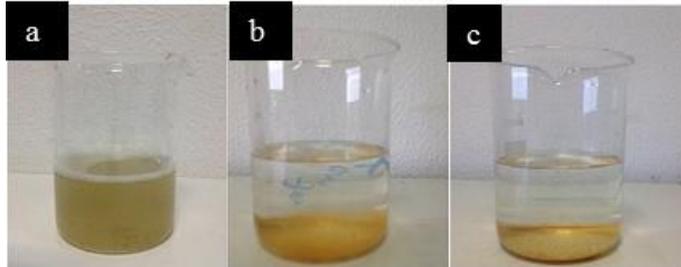


Fig. 5. Sample 1 collected on May 4, 2015 prior to Fenton and photo-Fenton treatment (a), sample 1 after Fenton treatment (b), and after photo-Fenton treatment (c).

The appearance of the crude sample was modified for the treated samples, and the parameters were analysed.

3.1. Physical parameters of the effluent: Turbidity and color

Turbidity and color values for samples 1 and 2 of the car washing effluent are shown in Table 1. The analyses were performed before and after undergoing Fenton and photo-Fenton treatment.

Table 1. Results of turbidity and color analysis for sample 1 and 2 before and after undergoing Fenton and photo-Fenton treatment.

Sample	Turbidity (UNT)		Color (uH)	
	1	2	1	2
Crude effluent	153.5	146.9	80	80
Fenton	36.4	33.3	20	10
Photo-Fenton	10.2	6.9	10	10
CONAMA 357 (2005) [26]	100		75	

As shown in Table 1, turbidity and color values were reduced for the two samples by both the Fenton and photo-Fenton processes. Sample 1 showed a 76.28% reduction in turbidity after Fenton treatment, whereas after photo-Fenton treatment, the reduction was 93.35%. Sample 2 showed a 77.34% reduction in turbidity after Fenton treatment and a 95.26% reduction in turbidity after photo-Fenton treatment. Both treatment processes reduced the turbidity rate to values below the maximum wastewater discharge standard, which is 100 UNT, according to the CONAMA Resolution 357 (2005) [43]. The photo-Fenton

treatment was more efficient compared to the Fenton treatment, but both processes showed a significant reduction in the presence of organic matter.

The samples collected from the crude effluent of car wash had 80 uH color as shown in Table 1. Sample 1 showed a 75.0% reduction in color after Fenton treatment, whereas after photo-Fenton treatment, the color reduction was 87.5%. Sample 2 had an 87.5% reduction in color after both processes. The color of wastewater does not pose a risk to health; its reduction indicates low content of iron and manganese, and complexes such as humic and fulvic acids (75% to 85% of cases). However, color of water that exceeds 15 uH can be perceived in a glass of water.

3.2. Chemical parameters of the effluent: Hydrogen ionic potential, surfactants, oils and greases, and chemical demand of oxygen

The pH was adjusted according to the requirements for Fenton and photo-Fenton reactions. The samples of crude effluents had pH within the legal parameters. In the state of Santa Catarina, according to the State Law N.º 14675, as of April 13, 2009, pH values for wastewater disposal should range from 6 to 9 [44]. The national parameter [6] requires a pH range between 5 and 9, which means that the state law is more restrictive than the national law.

Figures 6, 7 and 8 show the results of surfactants, oils and greases, and chemical oxygen demand for samples 1 and 2, before and after undergoing Fenton and photo-Fenton treatment.

Figure 6 exhibits the results of surfactants for samples 1 and 2, before and after undergoing Fenton and photo-Fenton treatment and also displays the parameter values for wastewater disposal, according to current legislation [27].

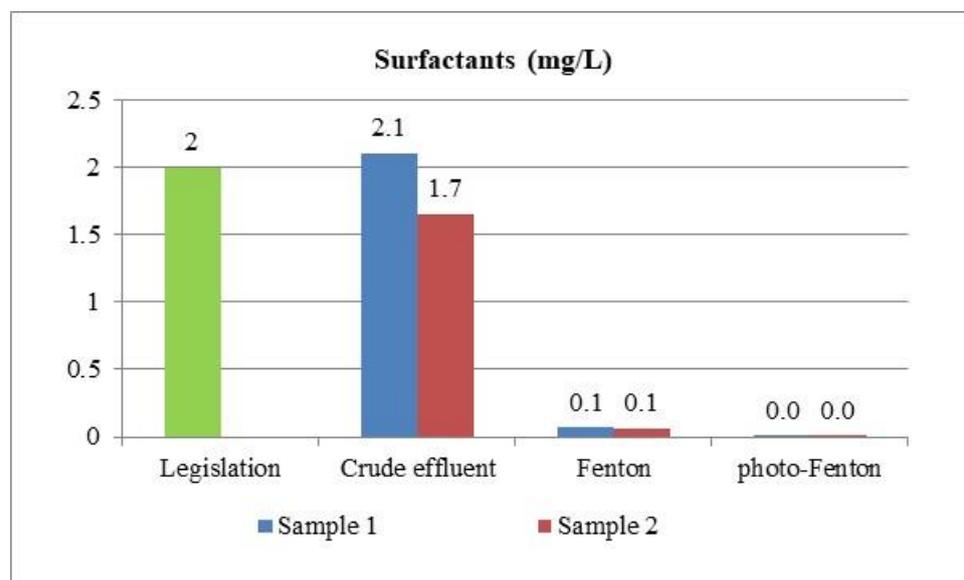


Fig. 6. Results of the surfactant analysis for samples 1 and 2 before and after undergoing Fenton and photo-Fenton treatment.

Before Fenton and photo-Fenton reactions, sample 1 had 2.1 mg/L and sample 2 had 1.65 mg/L of surfactants (Fig. 6). After undergoing Fenton treatment, sample 1 showed a 96.7% reduction in surfactant concentration, whereas as after the photo-Fenton treatment, a 99.5% reduction was obtained. For sample 2, the surfactant reduction was 96.4% after the Fenton treatment and 99.9% after the photo-Fenton treatment.

According to the state Law N.º 14675 (2009) [44], the maximum surfactant concentration for wastewater disposal should be 2 mg/L. Thus, the crude effluent in sample 2 could be released into water bodies without any further treatment. However, the reduction of this parameter was evaluated by oxidative processes, which showed a reduction in the levels of surfactant concentration, thus reducing the environmental risk of this effluent. A potential problem lies in foam formation and the likelihood of nitrogen and phosphorus emissions. These ingredients can persist in lakes and streams, and could create algae and cause eutrophication [17, 28]. Another possibility is that surfactants may create a bacterial population increase, transmitting through the food chain to protozoa, which are more sensitive to car wash toxins [29]. Detergents may damage mucous membranes and gills of fish to some extent. The gills may lose natural oils, thus interrupting oxygen transfer [30].

Figure 7 exhibits the results of oil and grease analysis for samples 1 and 2, before and after undergoing Fenton and photo-Fenton treatment and also displays the parameter values for wastewater disposal, according to current legislation [6].

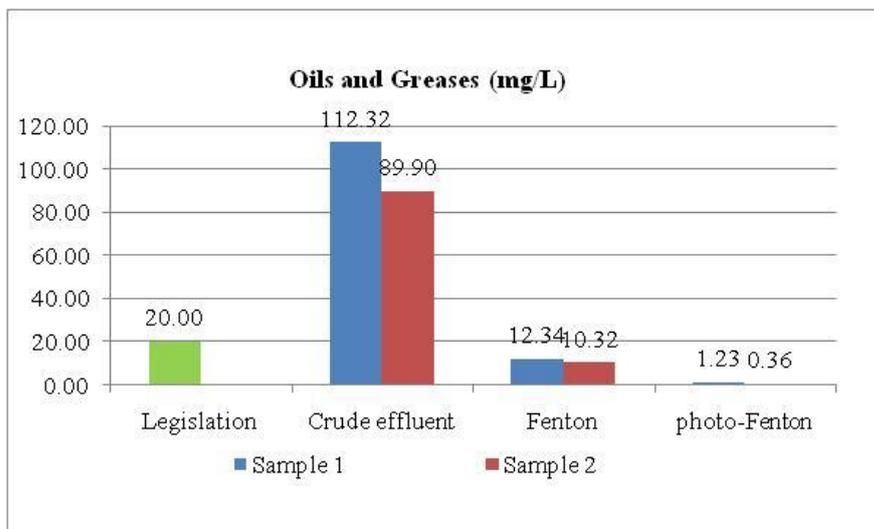


Fig. 7. Results of the oil and grease analysis for samples 1 and 2 before and after undergoing Fenton and photo-Fenton treatment.

In determining the oil and grease concentration, no emulsification occurred, given the low concentration of surfactants (Fig. 7). The crude effluent presented a result of 112.32 mg/L of oil and grease concentration for sample 1, and 89.8 mg/L for sample 2. The reduction for sample 1 after undergoing the Fenton treatment was 89.0%, whereas after the photo-Fenton treatment, there was a 98.9%

reduction in oil and grease concentrations. In sample 2, there was an 88.5% reduction in oil and grease concentration after the Fenton treatment and a 99.6% reduction after the photo-Fenton process. The results obtained were below the maximum value of oil and grease concentrations allowed for wastewater disposal (20 mg/L), according to the CONAMA resolution 430/2011, and photo-Fenton was the most efficient treatment [7].

The reduction in oil and grease concentration is vital, because besides depleting the aesthetic value, it can also compromise the oxygen content dissolved in water [29, 45].

Figure 8 exhibits the results of chemical oxygen demand for samples 1 and 2, before and after undergoing Fenton and photo-Fenton treatment.

The chemical oxygen demand before Fenton and photo-Fenton reactions presented a COD value of 201.43 mg/L for sample 1, and 133.44 mg/L for sample 2 (Fig. 8). For sample 1, there was an 83.9% reduction of COD after Fenton treatment, and a 93.9% reduction after photo-Fenton treatment. For sample 2, COD reduction was 83.3% after Fenton treatment and 92.3% after photo-Fenton treatment. Because it is an indirect measurement parameter, COD values are directly related to the values of suspended solids, oils and greases, and surfactants. COD reduction indicates chemical oxidation, whether biodegradable or not. The state legislation of Santa Catarina [44] and CONAMA Resolutions N.º 357 [42] and 430 [7] do not stipulate maximum concentration standards for this parameter in the recipient water bodies.

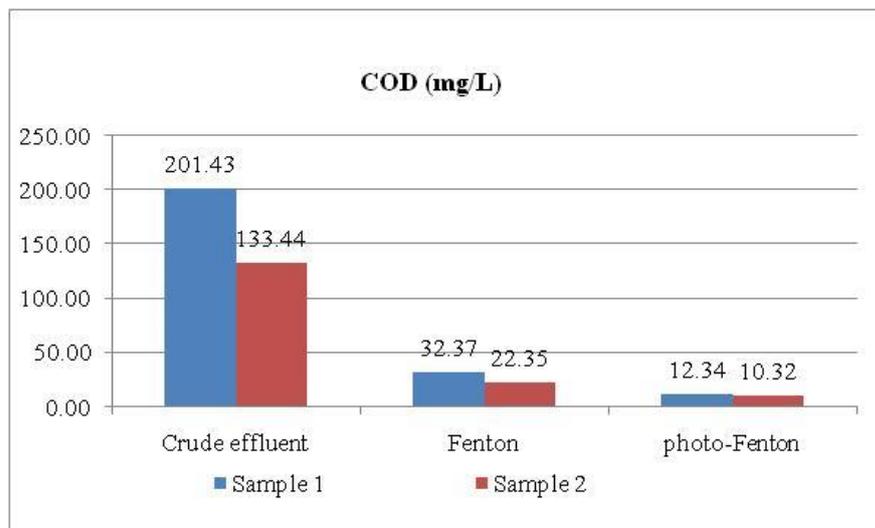


Fig. 8. Results of the chemical oxygen demand analysis for samples 1 and 2 before and after undergoing Fenton and photo-Fenton treatment.

4. Conclusions

The samples of residual wastewater from car washing submitted to Fenton and photo-Fenton oxidative processes removed the parameters of color, turbidity, surfactants, chemical oxygen demand, and oils and greases. The effluent had a reduction of these parameters between 75.0% and 99.6%, showing the efficiency of the Fenton and photo-Fenton processes.

The implementation of increasingly stringent regulations for wastewater disposal reinforces research efforts for the implementation of new treatments or for the improvement of those currently available. Future studies should investigate further on the effect of treatment time for wastewater cleaning. The global decline in water resources is forcing policy makers to impose regulations on freshwater use, as well as water reuse and recycling.

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